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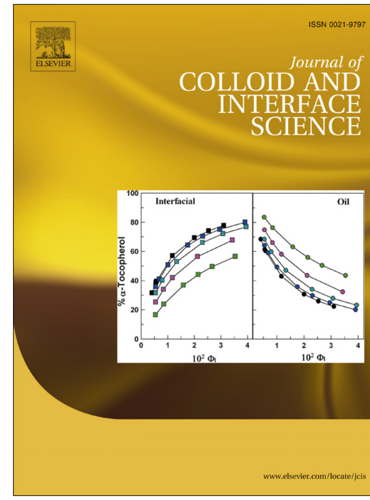

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Microfluidic Generation of Uniform Water Droplets Using Gas as the Continuous Phase

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Abstract: Microfluidic schemes for forming uniform aqueous microdroplets usually rely on contacting the aqueous liquid (dispersed phase) with an immiscible oil (continuous phase). Here, we demonstrate that the oil can be substituted with gas (nitrogen or air) while still retaining the ability to generate discrete and uniform aqueous droplets. Our device is a capillary co-flow system, with the inner flow of water getting periodically dispersed into droplets by the external flow of gas. The droplet size and different formation modes can be tuned by varying the liquid and gas flow rates. Importantly, we identify the range of conditions that correspond to the "dripping mode", i.e., where discrete droplets are consistently generated with no satellites. We believe this is a significant development that will be beneficial for chemical and biological applications requiring clean and contaminant-free droplets, including DNA amplification, drug encapsulation, and microfluidic cell culture.

Introduction

Conventional droplet microfluidics involves the formation of discrete microscale droplets within microchannels.[1,2] Droplets generated by microfluidics tend to be very uniform in size, and their high surface-to-volume ratios allow for rapid diffusion and heat transfer with their surroundings.[3] Accordingly, such droplets have been explored as microreactors or assay containers,[4,5] templates for particle synthesis,[6] cell carriers,[7,8] and even computing logic units.[9] Droplets are usually formed by contacting two immiscible liquid streams within the device: e.g., a stream of water (dispersed phase, DP) and a stream of oil (continuous phase, CP). At the meeting point of the streams (e.g., at a T-junction), the CP induces shear forces that pinch off the DP into individual droplets. The resulting aqueous droplets are transported within the oily CP, with surfactants in the CP stabilizing the interface and thus preventing coalescence of adjacent droplets.[2,3]

Thus far, the use of a liquid as *both* continuous and dispersed phases is considered foundational in the field of droplet microfluidics. However, several limitations arise due to the use of immiscible liquids. For example, in the case where aqueous droplets are crosslinked into microparticles or microgels, the particles still need to be washed to remove residual oil from their surfaces. Often, multiple washing steps are required, and these steps almost always have to be conducted off-chip. This is especially problematic if the particles are employed to encapsulate biological payloads such as cells or proteins—the washing steps can then negatively impact the viability of the encapsulants. In other cases, the water/oil interface constrains the utility of the on-chip droplets. For example, during DNA amplification within droplets via the polymerase chain reaction (PCR), it is known that the key enzyme involved in polymerizing DNA (Taq polymerase) irreversibly binds and denatures at the water/oil interface, thus reducing the efficiency of amplification.[5] Moreover, surfactants that are added to the continuous phase for droplet stabilization can act as a source of contamination, with adverse effects for biological and chemical assays.[3]

In this paper, we investigate an alternative to conventional droplet microfluidics, in which gas (nitrogen or air) is used to replace oil as the CP fluid. In this water-in-gas (W/G) approach, aqueous droplets are generated within a gaseous continuous phase, thereby ensuring that the droplets are inherently free of oil and surfactant contamination. Our focus is on establishing a microfluidic W/G platform that offers robust and repeatable operation, stable generation of individual droplets at periodic intervals, and precise control of droplet size. Towards this end, we have constructed a device based on concentric glass capillaries with their inner surfaces rendered hydrophobic. Water (DP) is sent through the inner capillary while gas (CP) flows through the outer surrounding capillary. At the meeting point of the streams, uniform water droplets are produced, with the size and periodicity of the droplets controlled by the water and gas flow rates. The droplets remain intact while traveling downstream, allowing them to be further manipulated or collected for analysis. We find that discrete droplets with no satellites are produced for a set of flow rates, and we map out the different droplet formation regimes.

To our knowledge, W/G droplet generation within microfluidic devices has not yet been studied systematically, and is not currently used by most researchers. This is possibly due to a lack of systematic guidelines for producing uniform droplets in W/G setups. It should be noted, though, that the dispersion of liquid streams into droplets using air has been widely studied for applications in ink-jet printers, sprayers and atomizers.[10-12] However, the droplets in those cases are dispersed into open environments, not in microchannels. Also, processes like atomization produce a wide distribution of droplet sizes rather than a uniform population. More recently, a few studies have been published on contacting water and gas